EFFECT OF γ-IRRADIATION ON THE PHOTOLUMINESCENCE OF $Cd_{1-x}Zn_xTe$ CRYSTALS

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We study the effect of γ-irradiation (with doses in the range 10–100 kGy) on the low-temperature ($T = 5$ K) photoluminescence of $Cd_{1-x}Zn_xTe$ ($x = 0.05$) crystals. The following phenomena induced by γ-irradiation were observed: a) essential decrease of the intensities of the initial (as-grown) luminescence bands – the defect band ($h\nu_m = 1.409$ eV), these caused by donor-acceptor pairs ($h\nu_m = 1.547$ eV) and shallow acceptors ($h\nu_m = 1.556$ eV), as well as bands related to excitons bound to shallow neutral acceptors and donors ($h\nu_m = 1.592$ eV and $h\nu_m = 1.599$ eV, respectively), which results from the decrease of the concentration of the corresponding luminescence centers due to their interaction with radiation-induced defects; b) appearance of new luminescence bands probably caused by radiation-induced cadmium vacancies V_{Cd} bound to other defects (donor-acceptor pairs, $h\nu_m = 1.548$ eV) and isolated cadmium vacancies ($h\nu_m = 1.557$ eV) as well as excitons bound to the indicated cadmium vacancies ($h\nu_m = 1.590$ eV). The intensity of the radiation-induced bands changes nonmonotonically with increase in the dose of γ-irradiation: it firstly grows at low Φ_γ (≤ 50 kGy) due to an increase of the concentration of cadmium vacancies and then considerably decreases at high $\Phi_\gamma > 50$ kGy due to the generation of a large number of effective centers of radiationless recombination of excess charge carriers.

1. Introduction

Investigations of the effect of high-energy particles on electric properties of intermetallic semiconductors allow one to obtain the important information on the generation of radiation-induced defects, their physical properties, and interaction of these defects with those present in a non-irradiated material (see, for example, CdTe [1,2] and $Cd_{1-x}Zn_xTe$ [3–5]). This work presents data on the influence of γ-irradiation on the low-temperature ($T = 5$ K) photoluminescence of $Cd_{1-x}Zn_xTe$ ($x = 0.05$) crystals. It is shown that γ-irradiation results in significant changes of the photoluminescence of the investigated crystals, which is caused by the appearance of radiation-induced defects (cadmium vacancies). The regularities observed in the case of such irradiation are explained.

2. Experimental Technique

Our investigations were performed for p-$Cd_{1-x}Zn_xTe$ crystals [with the room-temperature resistivity $\rho \approx 60$ Ohm·cm and $\rho \to \infty$ at $T = 5$ K (the conduction of the crystals at $T = 5$ K is determined by excess electrons and holes)]) grown with the help of the Bridgman technique. Single-crystal $Cd_{1-x}Zn_xTe$ plates with (111) orientation and a thickness of 1–1.5 mm were cut from ingots 40 mm in diameter. In order to remove the damaged surface layer 150–200 μm in thickness, the surface of the cut plates was treated by means of the chemical-mechanical polishing in a bromine-methanol etchant [6]. The content of zinc in the studied samples was determined using the X-ray diffractometry technique ($x = 0.05$). The crystals were irradiated by a ^{60}Co source (with a quantum energy of 1.2 MeV) in the dose range 10–100 kGy (the γ-quantum flux $N_\gamma = 1.69 \times 10^{15} - 1.69 \times 10^{16}$ quanta/cm^2) at room temperature. The photoluminescence spectra of the investigated p-$Cd_{1-x}Zn_xTe$ crystals were studied at the temperature $T = 5$ K. The luminescence was excited by an LGN-222 He–Ne laser (at the excitation intensity $L = 10^{19}$ quanta/(cm^2·s), the quantum energy equaled 1.96 eV). The low-temperature photoluminescence spectra in the range 1.3–1.7 eV were obtained with the help of an MDR-23 monochromator, while the signal was registered by a cooled FEU-62.

3. Results and Discussion

Figure 1 shows the low-temperature ($T = 5$ K) photoluminescence spectra of the $Cd_{1-x}Zn_xTe$ ($x =$

0.05) crystals: initial one and those irradiated by various doses of γ-quanta ($\Phi_\gamma \leq 100$ kGy). Analyzing the observed luminescence bands (their intensities I, positions of maxima $h\nu_m$, and half-widths w), we can make the following conclusions:

1) the luminescence spectra of the initial crystals contain the following bands: a) a luminescence band with $h\nu_m = 1.409$ eV and $w = 74$ meV caused by the so-called A-centers (the latter form deep levels that include cadmium vacancies $V_{\rm Cd}$ and various uncontrolled donor impurities) hereinafter called a defect band D [7–11]; b) a complex (nonelementary) luminescence band with $h\nu_m = 1.550$ eV and $w = 20.3$ meV ($D^0A^0 + eA^0$ band) caused (as is known from [1,2,7,9]) by the recombination in donor-acceptor pairs D^0A^0 formed by shallow neutral donors D^0 and acceptors A^0 (hereinafter called a D^0A^0 band with $h\nu_m = 1.547$ eV and $w = 13.5$ meV) and transitions of free electrons e to shallow neutral acceptors A^0 (eA^0 band with $h\nu_m = 1.556$ eV and $w = 13.5$ meV); the ratio of their intensities is equal to 1.7, i.e. the emission induced by the donor-acceptor recombination dominates in the complex band (see Fig. 2,*a*)[1]; c) luminescence bands with $h\nu_m = 1.592$ eV and $w = 8$ meV and $h\nu_m = 1.599$ eV and $w = 2.9$ meV caused by the annihilation of excitons X bound to shallow neutral acceptors A^0(A^0X band) and shallow neutral donors D^0 (D^0X bands), respectively [1, 2, 7, 9].

2) γ-irradiation of the $Cd_{1-x}Zn_xTe$ crystals results in the following effects: a) considerable decrease of the intensities of the initial bands (the defect band D and the exciton bands A^0X and D^0X) and the complex band $D^0A^0 + eA^0$, i.e. the intensities of the elementary bands D^0A^0 and eA^0; this phenomenon is caused the significant reduction of the concentration of the initial D, D^0A^0, eA^0, A^0X, and D^0X luminescence centers under γ-irradiation due to the radiation-induced conversion of the latter to other centers with a different structure (more simple or complex) resulting from their interaction with radiation-induced defects [1,2,12,13]; b) appearance of new intense luminescence bands differing from those characteristic of the initial samples in the positions of luminescence maxima (their intensities considerably exceed those of the initial luminescence bands with a similar scheme of constituent radiative transitions): b1) a complex (two-component) luminescence band with $h\nu_m = 1.549$ eV and $w = 16.2$ meV (hereinafter called a $D^*V^0_{\rm Cd} + eV^0_{\rm Cd}$ band) caused by the recombination in donor-acceptor pairs $D^*V^0_{\rm Cd}$ formed by initial defects (or impurities) of the donor nature D^*, radiation-induced neutral cadmium vacancies $V^0_{\rm Cd}$ ($D^*V^0_{\rm Cd}$, this band is characterized by $h\nu_m = 1.548$ eV and $w = 13.6$ meV), and transitions of free electrons to shallow neutral acceptors $V_{\rm Cd}$ formed due to irradiation ($eV^0_{\rm Cd}$ band with $h\nu_m = 1.557$ eV and $w = 13.5$ meV). The intensity of the $D^*V^0_{\rm Cd}$

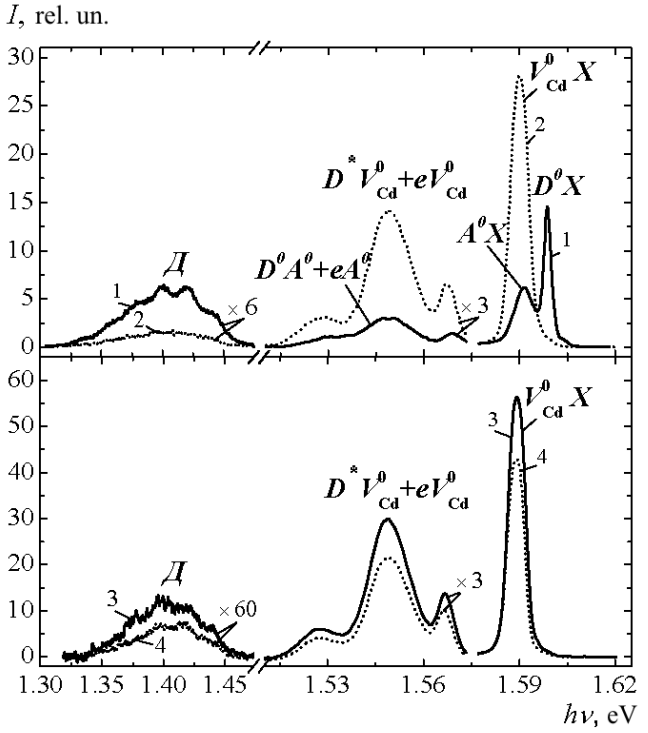

Fig. 1. Photoluminescence spectra of the initial (1) and γ-irradiated $Cd_{1-x}Zn_xTe$ ($x = 0.05$) crystals. $T = 5$ K, $\Phi_\gamma = 10$ (2), 50 (3), and 100 kGy (4). The spectra show the true ratios between the band intensities in different crystals

[1] The data on the characteristics (I, $h\nu_m$, and w) of the D^0A^0 and eA^0 elementary luminescence bands presented hereinafter were obtained by means of the decomposition of the $D^0A^0+eA^0$ complex band into the elementary components D^0A^0 and eA^0 and their LO-phonon replicas (corresponding to the emission of a longitudinal optical phonon LO with an energy of 22 meV) D^0A^0– LO ($h\nu_m = 1.525$ eV and $w = 13.5$ meV), eA^0– LO ($h\nu_m = 1.534$ eV, $w = 13.5$ meV, and $\bar{S} = 0.33$) and A^0X – LO ($h\nu_m = 1.570$ eV, $w = 8$ meV, and $\bar{S} = 0.55$), see Fig. 2,it a [1, 2, 9, 14] {here, $\bar{S}$ is the average number of longitudinal optical phonons corresponding to a single act of radiationless recombination (the so-called Huang–Rhys factor) determined by the ratio of the intensities of the *1LO*-phonon replica and the corresponding zero-phonon band [1,2,14]}. The decomposition is performed taking into account that the eA^0 band partially overlaps with the LO -phonon replica of the A^0Xband ($h\nu_m = 1.570$ eV, $w = 8$ meV, and $\bar{S} = 0.21$), see Fig. 2,*a*.

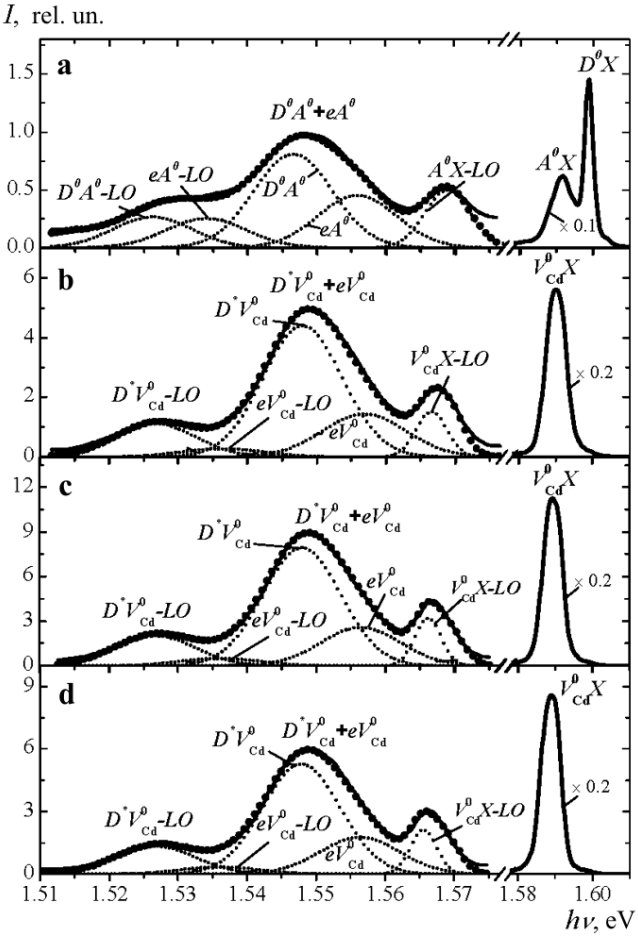

Fig. 2. Decomposition of the complex bands $D^0A^0 + eA^0$ and $D^*V^0_{\rm Cd}$+$eV^0_{\rm Cd}$ observed in the initial (*a*) and γ-irradiated ($\Phi_\gamma = 10$ (*b*), 50 (*c*), and 100 kGy (*d*)) $Cd_{1-x}Zn_xTe$ ($x = 0.05$) crystals into the elementary D^0A^0, eA^0 and $D^*V^0_{\rm Cd}$, $eV^0_{\rm Cd}$ components and their *LO*-phonon replicas (at $h\nu_m \geq 1.56$ eV, the emission of the $D^0A^0 + eA^0$ and $D^*V^0_{\rm Cd} + eV^0_{\rm Cd}$ complex bands overlaps with the luminescence of the *LO*-phonon replica of the A^0X band caused by excitons bound to shallow acceptors). The decomposition is performed under the assumption that the indicated luminescence bands are described by Gaussian curves (see the text) [1, 2, 9, 14]. Dots show the theoretical complex spectra $D^0A^0 + eA^0$ and $D^*V^0_{\rm Cd} + eV^0_{\rm Cd}$ determined by a sum of the theoretical elementary bands D^0A^0, eA^0 and $D^*V^0_{\rm Cd}$, $eV^0_{\rm Cd}$ and their (and those of the A^0X band) phonon replicas (see the text) [14]. The spectra show the true ratios between the band intensities in the initial and irradiated crystals

band exceeds that of the $eV^0_{\rm Cd}$ one approximately threefold, i.e. the dominant emission in the complex band is created by the donor-acceptor recombination (see Fig. 2,*b–d*)[2]; b2) a luminescence band with $h\nu_m = 1.589$ eV and $w = 6.5$ meV formed due to the annihilation of bound excitons $V^0_{\rm Cd}X$ created by radiation-induced neutral cadmium vacancies $V^0_{\rm Cd}$ and excitons X ($V^0_{\rm Cd}X$ band)[3].

The radiation-induced luminescence bands in the studied crystals are assumed to be formed by centers that include radiation-induced neutral cadmium vacancies $V^0_{\rm Cd}$ (cadmium vacancies form shallow acceptors in CdTe [1,2] and most probably in the studied crystals as well), i.e. we suppose that the appearance of radiation-induced luminescence bands is related to the latter. A rather high probability of this assumption is confirmed by the following facts: a) the concentration of radiation-induced cadmium vacancies in irradiated crystals is somewhat higher than the concentration of tellurium vacancies [10,11] (the latter induce a luminescence band with $h\nu_m = 1.1$ eV in $Cd_{0.9}Zn_{0.1}Te$ at $T = 4.2$ K [10], i.e., a luminescence band significantly differing from that observed in irradiated crystals); it also exceeds the concentration of zinc vacancies (which is due to a rather low content of zinc in the investigated $Cd_{0.95}Zn_{0.05}Te$ crystals); b) it is worth expecting that radiation-induced interstitial atoms of the $Cd_{0.95}Zn_{0.05}Te$ lattice (isolated ones and bound to impurities or other defects) do not make a considerable contribution to the observed γ-induced luminescence as follows from the known positions of some levels they create [3, 10] (they considerably differ from the corresponding ones for the observed γ-induced luminescence centers). When identifying the scheme of electron transitions resulting in the appearance of the radiation-induced luminescence bands with $h\nu_m = 1.548$, 1.557, and 1.590 eV, we took into account that the indicated emission energies $h\nu_m$ are characteristic of the luminescence caused by the recombination in donor-acceptor pairs, transitions of free electrons to shallow neutral acceptors, and annihilation of excitons

[2] The forms of the indicated $D^*V^0_{\rm Cd}$ and $eV^0_{\rm Cd}$ elementary luminescence bands obtained by means of the decomposition of the $D^*V^0_{\rm Cd} + eV^0_{\rm Cd}$ complex band into elementary components and their *LO*-phonon replicas $D^*V^0_{\rm Cd}$–*LO* ($h\nu_m = 1.526$ eV, $w = 13.6$ meV, and $\bar{S} = 0.23$) and $eV^0_{\rm Cd}$ – *LO* ($h\nu_m = 1.535$ eV and $w = 13.5$ meV, and $\bar{S} = 0.2$) are shown in Fig. 2,*b–d* [1,2,9,14]. It is taken into account that the $eV^0_{\rm Cd}$ band partially overlaps with the *LO* phonon replica of the $V^0_{\rm Cd}X$ band ($h\nu_m = 1.568$ eV, $w = 6.5$ meV, and $\bar{S} = 0.06$). This decomposition allowed us to determine the main characteristics (I, $h\nu_m$, w, and $\bar{S}$) of the $D^*V^0_{\rm Cd}$ and $eV^0_{\rm Cd}$ elementary bands and their phonon replicas.

[3] For the $D^*V^0_{\rm Cd}$, $eV^0_{\rm Cd}$, and $V^0_{\rm Cd}X$ radiation-induced bands, $h\nu_m$, $w \neq f(\Phi_\gamma)$, see below.

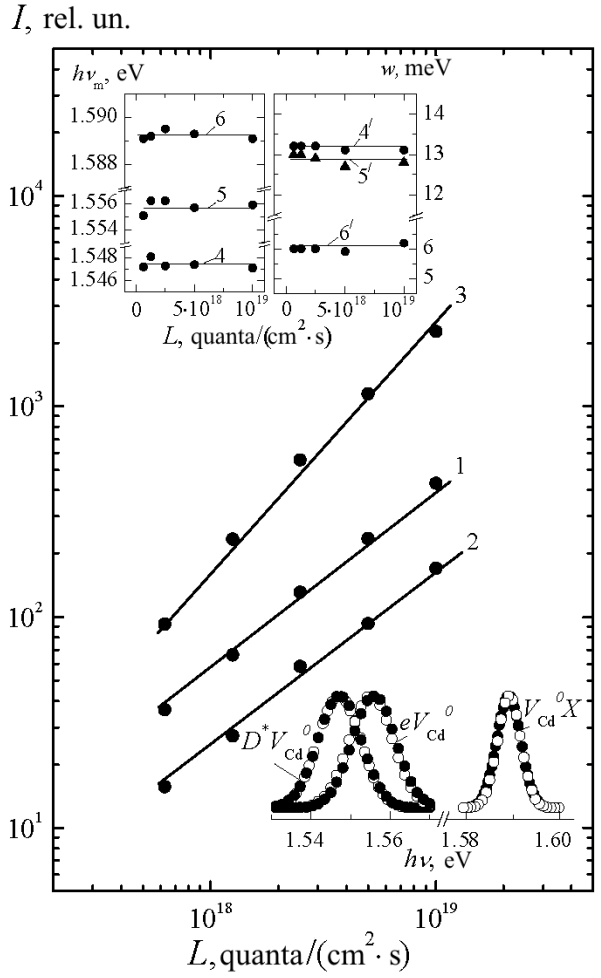

Fig. 3. Low-temperature ($T = 5$ K) intensities of the $D^*V_{Cd}^0$ (*1*), eV_{Cd}^0 (*2*), and V_{Cd}^0X (*3*) radiation-induced luminescence bands as functions of the excitation intensity L in the $Cd_{1-x}Zn_xTe$ ($x = 0.05$) crystals irradiated by γ-quanta ($\Phi_\gamma = 10$ kGy). In the experiment, $I \sim L^{0.8}$ (*1, 2*), $L^{1.3}$ (*3*). The curves show the true ratios between the intensities of the radiation-induced luminescence bands. The figure also shows the positions of maxima (*4–6*) and half-widths (*4′–6′*) of the $D^*V_{Cd}^0$ (*4, 4′*), eV_{Cd}^0 (*5, 5′*), and V_{Cd}^0X (*6, 6′*) radiation-induced luminescence bands for the γ-irradiated crystal at $T = 5$ K. The inset presents the forms of the $D^*V_{Cd}^0$, eV_{Cd}^0, andV_{Cd}^0X radiation-induced luminescence bands normalized to the emission maximum at $L = 5 \times 10^{17}$ (○) and 10^{19} (●) quanta/cm^2·s observed in the considered γ-irradiated crystals at $T = 5$ K

bound to shallow neutral acceptors, respectively [1, 2, 7, 9, 14].

It is worth noting the following facts. First, the intensities of the $D^*V_{Cd}^0$ and eV_{Cd}^0 radiation-induced bands grow sublinearly [$I(D^*V_{Cd}^0)$, $I(eV_{Cd}^0) \sim L^{0.8}$), and that

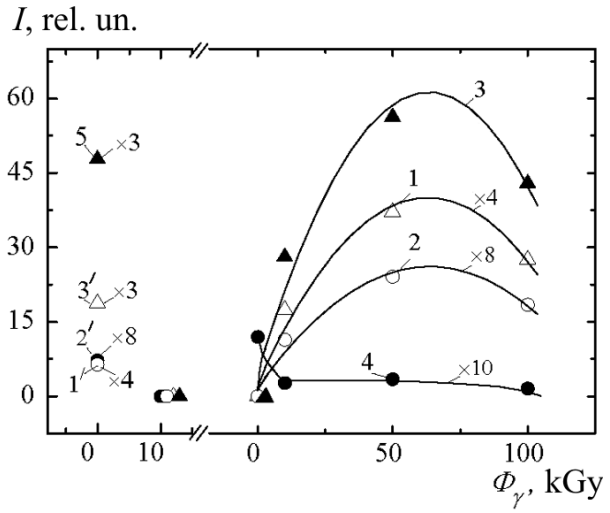

Fig. 4. Dose dependences of the low-temperature ($T = 5$ K) intensities of the radiation-induced $D^*V_{Cd}^0$ (*1*), eV_{Cd}^0 (*2*), and V_{Cd}^0X (*3*) and the initial D (*4*) luminescence bands observed in the γ-irradiated $Cd_{1-x}Zn_xTe$ ($x = 0.05$) crystals. The figure also shows the intensities of the initial D^0A^0 (*1′*), eA^0 (*2′*), A^0X (*3′*), and D^0X (*5*) luminescence bands in the studied crystals at $\Phi_\gamma = 0$ and 10 kGy. The dependences I (Φ_γ) show the true ratios between the intensities of luminescence bands in the investigated crystals

of the V_{Cd}^0X one – superlinearly [$I(V_{Cd}^0X) \sim L^{1.3}$] with increase in the excitation intensity L (the forms of the indicated radiation-induced luminescence bands, particularly, the positions of their maxima and half-widths, do not depend on L). Thus, as expected [9,17,18], the intensity of the V_{Cd}^0X band changes with L much stronger than the intensities of the $D^*V_{Cd}^0$ and eV_{Cd}^0 ones, see Fig. 3. Second, a relatively large half-width of the eV_{Cd}^0 band (as well as that of the eA^0 one, see above) is probably caused by the strong interaction of the eV_{Cd}^0 (and eV^0) luminescence centers with acoustic phonons, i.e. the strong electron-acoustic phonon coupling in the indicated centers (this fact results in the Gaussian form of the eV_{Cd}^0 and eA^0 luminescence bands) [15] {$w = 1.8\,kT$ (here, k is the Boltzmann constant), i.e. it is rather small at low temperatures ($w = 0.75$ meV at $T = 5$ K), if the luminescence band is determined by phonon-less radiative transitions of free electrons to acceptors [16, 19]}.

Figure 4 presents the intensities of the $D^*V_{Cd}^0$, eV_{Cd}^0, and V_{Cd}^0X radiation-induced bands as functions of the irradiation dose Φ_γ. One can see that the intensity of these bands firstly significantly increases (approximately equally) with increase in the radiation dose at low Φ_γ

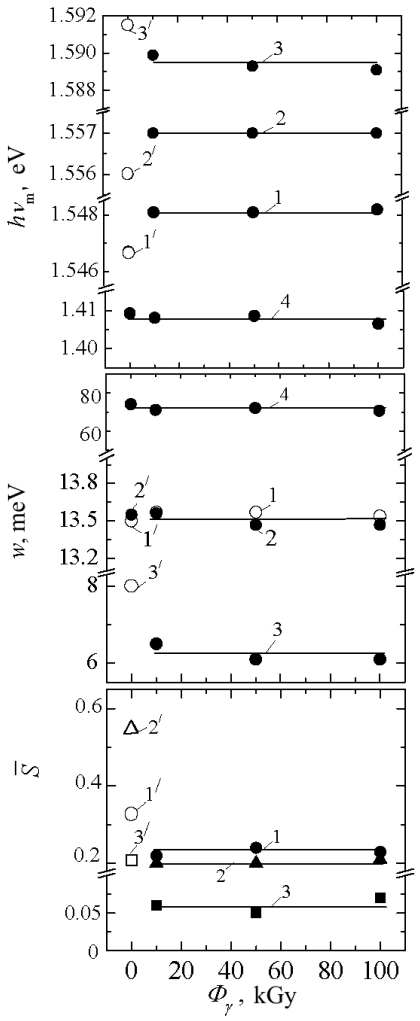

Fig. 5. Dose dependences of the maximum positions $h\nu_m$, half-widths w, and average numbers of phonons for the radiation-induced $D^*V_{\rm Cd}^0$ (*1*), $eV_{\rm Cd}^0$ (*2*), and $V_{\rm Cd}^0X$ (*3*) and initial D (*4*) luminescence bands observed in the γ-irradiated $Cd_{1-x}Zn_xTe$ ($x = 0.05$) crystals at $T = 5$ K. The values of $h\nu_m$, w, and $\bar{S}$ at $T = 5$ K for the initial D^0A^0 (*1′*), eA^0 (*2′*), and A^0X (*3′*) luminescence bands in the non-irradiated $Cd_{1-x}Zn_xTe$ crystals are also given ($\Phi_\gamma = 0$)

and then falls at high Φ_γ[4] (the position of the maximum and the half-width of the radiation-induced impurity and exciton luminescence bands weakly change with Φ_γ, see Fig. 5)[5]. The observed growth of the intensities of the $D^*V_{\rm Cd}^0$, $eV_{\rm Cd}^0$, and $V_{\rm Cd}^0X$ bands with increase in the dose of γ-irradiation is caused by a rise in the concentration of radiation-induced cadmium vacancies (which is confirmed by the same form of the dose dependences of the indicated luminescence bands observed at low Φ_γ). The reduction of the intensities of the $D^*V_{\rm Cd}^0$ and $eV_{\rm Cd}^0$ bands at high Φ_γ is due to the generation of centers of radiationless recombination of excess carriers under γ-irradiation (the rate of recombination of charge carriers via these centers ultralinearly grows with increase in Φ_γ), which results in a considerable reduction of the concentration of the latter. The fall in the intensity of the $V_{\rm Cd}^0X$ exciton band with increase in Φ_γ is caused by a decrease of the concentration of free excitons resulting from a reduction of the concentrations of excess electrons and holes. As was noted above, the latter phenomenon is explained by the raised recombination of electrons and holes at radiation-induced effective centers of radiationless recombination of charge carriers.

The value of $\bar{S}$ for the $D^*V_{\rm Cd}^0$, $eV_{\rm Cd}^0$, and $V_{\rm Cd}^0X$ radiation-induced luminescence bands (it is independent of the dose of γ-irradiation) is much lower than 1 (see Fig. 5), i.e. the electron-LO-phonon coupling in the indicated radiation-induced centers is weak [16]. Comparing the values of $\bar{S}$ for the radiation-induced and initial luminescence bands formed according to a similar scheme of radiative transitions, one can see that they are lower in the former case, that is $\bar{S}(D^*V_{\rm Cd}^0) < S(D^0A^0)$, $\bar{S}(eV_{\rm Cd}^0) < S(eA^0)$, and $\bar{S}(V_{\rm Cd}^0X) < S(A^0X)$, see Fig. 5. This implies that the electron-LO-phonon coupling in the $D^*V_{\rm Cd}^0$, $eV_{\rm Cd}^0$, and $V_{\rm Cd}^0X$ radiation-induced centers is weaker than that in the initial D^0A^0, eA^0, and A^0X ones, respectively.

The intensity of the defect band D considerably decreased by initial γ-irradiation ($\Phi_\gamma = 10$ kGy) weakly changes with the further growth of the dose Φ_γ (Fig. 4). This testifies that, at all Φ_γ, the given band is determined by the growth A-centers, whose composition can differ from that characteristic of the major number of A-centers (in particular, by the nature of uncontrolled donor impurities). This fact indicates the radiation stability of a part of these defects, i.e. a weak change of the concentration of a part of D defects under γ-irradiation (the D band is probably caused by defects of different nature [3, 4, 9]). As should be expected, the intensity

[4] Figure 4 also demonstrates the above-noted considerable decrease of the intensities of the initial D^0A^0, eA^0, A^0X, and D^0X bands under γ-irradiation, as well as the fact that the intensity of the $D^*V_{\rm Cd}^0$, $eV_{\rm Cd}^0$, and $V_{\rm Cd}^0X$ radiation-induced bands is much higher than that of the initial D^0A^0, eA^0, and A^0X ones, respectively, with a similar scheme of constituent radiative transitions.

[5] Figure 5 also illustrates the above-noted significant difference of the positions of maxima and half-widths of the $D^*V_{\rm Cd}^0$, $eV_{\rm Cd}^0$, and $V_{\rm Cd}^0X$ radiation-induced luminescence bands from those of the initial D^0A^0, eA^0, and A^0X ones with a similar scheme of constituent radiative transitions.

of this band decreases under irradiation by the maximal ($\Phi_\gamma = 100$ kGy) radiation flux due to the effective generation of centers of radiationless recombination of charge carriers at this irradiation dose.

4. Conclusions

The γ-irradiation of $Cd_{1-x}Zn_xTe$ crystals considerably changes the form of their low-temperature photoluminescence spectra. It results in a significant decrease of the intensity of the initial luminescence bands due to a reduction of the concentration of as-grown defects due their interaction with radiation defects, as well as due to the appearance of new luminescence bands probably created by radiation-induced cadmium vacancies. The analysis of the regularities that govern a change in the intensities of the low-temperature photoluminescence bands observed under γ-irradiation is important for understanding the processes of interaction of primary and radiation-induced defects now studied in intermetallic semiconductors, as well as for clarification of the reasons resulting in their different radiation stabilities.

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ВПЛИВ γ-ОПРОМІНЕННЯ НА ФОТОЛЮМІНЕСЦЕНЦІЮ КРИСТАЛІВ $Cd_{1-x}Zn_xTe$

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Вивчено вплив опромінення різними потоками γ-квантів (доза 10–100 кГр) на низькотемпературну ($T = 5$ K) фотолюмінесценцію кристалів $Cd_{1-x}Zn_xTe$ ($x = 0,05$). Спостерігали стимульовані γ-опроміненням: а) суттєве зменшення інтенсивностей вихідних (ростових) смуг люмінесценції – дефектної ($h\nu_m = 1,409$ еВ), зумовленої донорно-акцепторними парами ($h\nu_m = 1,547$ еВ), мілкими акцепторами ($h\nu_m = 1,556$ еВ) та зумовлених зв'язаними на мілких нейтральних акцепторах і донорах екситонами ($h\nu_m = 1,592$ еВ та $h\nu_m = 1,599$ еВ відповідно) внаслідок зменшення концентрації відповідних центрів люмінесценції завдяки їх взаємодії з радіаційни-

ми дефектами; б) появу нових смуг люмінесценції, зумовлених, ймовірно, радіаційно-стимульованими вакансіями кадмію V_{Cd}, зв'язаними з іншими дефектами ($h\nu_m = 1,548$ еВ) та ізольованими вакансіями кадмію ($h\nu_m = 1,557$ еВ), а також екситонами, зв'язаних на вказаних вакансіях кадмію ($h\nu_m = 1,590$ еВ). Інтенсивність радіаційно-стимульованих смуг немонотонно змінюється зі збільшенням дози γ-опромінення: спочатку зростає при низьких Φ_γ (≤ 50 кГр), завдяки збільшенню концентрації вакансій кадмію, а потім значно зменшується при високих Φ_γ (≥ 50 кГр), завдяки генерації значної кількості ефективних центрів безвипромінювальної рекомбінації надлишкових носіїв струму.